Minicolloque MMB16 ⓧ **Oral** ☐ **Poster**

Drying Dynamics in Films of Nanoparticle Suspensions and Polymer Solutions

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When a complex soft matter solution is dried, the interplay of solvent evaporation, fluid dynamics, diffusion, phoresis, capillarity, and mechanics of solute materials leads to rich, far-from-equilibrium phenomena. Recently we have been using large scale molecular dynamics (MD) simulations to study the drying process of colloid suspensions, polymer solutions, and their mixtures with the solvents modeled explicitly. For suspensions containing bidisperse nanoparticle mixtures, the counterintuitive "small-on-top" stratification, with an enrichment of the smaller particles at the receding liquid-vapor interface during fast drying, is observed and a state diagram of stratification is determined.[1] Results are compared to the predictions of several theoretical models based on diffusiophoresis. We show that for rapid solvent evaporation, evaporative cooling leads a thermal gradient in the drying solution and the thermophoretic response of nanoparticles to this gradient is size-dependent. As a result, thermophoresis can compete with diffusiophoresis and suppress the "small-on-top" stratification at extremely fast evaporation rates. On the basis of this observation, we demonstrate with MD simulations an approach for controlling stratification using various externally imposed thermal gradients.[2] We further show that stratification also occurs in a mixture of two liquids being rapidly dried, with the less volatile component concentrated near the evaporating front.[3] As a result, a liquid mixture can be used to separate and stratify a particle mixture if different particles have contrasting interactions with the liquid components.[3] Finally, we establish a new strategy of uniformly dispersing nanoparticles into a polymer matrix using fast solvent evaporation, based on the stratification behavior of polymer-particle mixtures in a far-from-equilibrium situation induced by rapid drying.[4]

1. Y. Tang, G. S. Grest, and S. Cheng, Langmuir **34** (24), 7161 (2018).
2. Y. Tang, G. S. Grest, and S. Cheng, Langmuir **35** (12), 4296 (2019).
3. B. Liu, G. S. Grest, and S. Cheng, manuscript in preparation (2021).
4. S. Cheng and G. S. Grest, ACS Macro Letters **5**, 694 (2016).